#### PATENT COOPERATION TREATY

#### PCT

#### **NOTIFICATION OF ELECTION**

(PCT Rule 61.2)

#### From the INTERNATIONAL BUREAU

Commissioner **US Department of Commerce** United States Patent and Trademark Office, PCT 2011 South Clark Place Room CP2/5C24 Arlington, VA 22202 **ETATS-UNIS D'AMERIQUE** 

in its capacity as elected Office

Date of mailing (day/month/year) 09 October 2001 (09.10.01)

International application No.

02 March 2000 (02.03.00)

PCT/US00/05458 International filing date (day/month/year)

Applicant's or agent's file reference M 6712 HST/NI

Priority date (day/month/year) 02 March 1999 (02.03.99)

**Applicant** 

KAWAGUCHI, Jun et al

1.	The designated Office is hereby notified of its election made:
	X in the demand filed with the International Preliminary Examining Authority on:
	23 June 2000 (23.06.00)
	in a notice effecting later election filed with the International Bureau on:
2.	The election X was
	was not
	made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).
	-
l	

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland

Authorized officer

Elisabeth KÖNIG

Facsimile No.: (41-22) 740.14.35

Telephone No.: (41-22) 338.83.38

### INTERNATION SEARCH REPORT



A. CLASSIFICATION OF SUBJECT MATTER  IPC(7) :C23C 22/07  US CL :148/262					
	to International Patent Classification (IPC) or to boo	h national classification and IPC			
B. FIE	LDS SEARCHED				
Minimum	documentation searched (classification system follow	ed by classification symbols)			
	148/262, 148/241; 205/111, 318, 189				
Documenta	tion searched other than minimum documentation to the	e extent that such documents are incl	uded in the fields searched		
Electronic de EAST	data base consulted during the international search (	name of data base and, where practi	cable, search terms used)		
	rms: zinc phosphate, phosphoric acid, nitric acid, s	ludge, sludging, nonsludging			
C. DOC	CUMENTS CONSIDERED TO BE RELEVANT				
Category*	Citation of document, with indication, where a	ppropriate, of the relevant passages	Relevant to claim No.		
Y	US 5,525,431 A (KANAMARU et al 14-40; col. 15, lines 25-55; col. 18, lines and 63-68; col. 21, lines 1-2; col. 22,	nes 19-51; col. 20, lines 29-	nes 1-10 32		
Y	Y US 5,503,733 A (SPECKMANN et al) 02 April 1996, abstract, col. 2, lines 50-63; col. 3, lines 45-50; col. 4, lines 29-33 and 47-49; col 5, lines 16-42; claim 1.				
Y	Y US 5,203,930 A (BLUMLHUBER et al) 20 April 1993, abstract, col. 2, lines 25-30; col. 3, lines 17-27; claim 1.				
X Furth	ner documents are listed in the continuation of Box (	C. See patent family anne	x.		
	ecial categories of cited documents:	"T" later document published after the	ne international filing date or priority		
to	cument defining the general state of the art which is not considered be of particular relevance	the principle or theory underlying	ng the invention		
"L" doc	lier document published on or after the international filing date cument which may throw doubts on priority claim(s) or which is ad to establish the publication date of another citation or other		e; the claimed invention cannot be unsidered to involve an inventive step ne		
special reason (as specified)  *O*  document referring to an oral disclosure, use, exhibition or other means		"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art			
"P" doc the	nument published prior to the international filing date but later than priority date claimed	*&* document member of the same patent family			
Date of the	actual completion of the international search	Date of mailing of the internationa	l search report		
26 APRIL	26 APRIL 2000 16 MAY 2000				
Commissior Box PCT					
Washington Facsimile No	ь, D.C. 20231 o. (703) 305-3230	ANDREW L OLTMANS			
	(	Telephone No. (703) 308-0661	1		



Inernational application No. PCT/US00/05458

		PC1/US00/US43	_		
C (Continua	tion). DOCUMENTS CONSIDERED TO BE RELEVANT				
Category*	egory* Citation of document, with indication, where appropriate, of the relevant passages				
ď	US 4,950,339 A (GEHMECKER et al) 21 August 1990 a col. 1, lines 63-68; col. 2, lines 1-4 and 51 and 65; col. 52, claim 1.	Relevant to claim No			
7	US 3,647,568 A (LARSON) 07 March 1972, col. 2, lines 30-50.		1-7		

919147 PATENT COOPERATION TREATY

## **PCT**

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#### INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION		of Transmittal of International amination Report (Form PCT/IPEA/416)		
M 6712 HST/N International application No.	International filing date (day/mor		Priority date (day/month/year)		
			02 March 1999 (02.03.1999)		
PCT/US00/05458 International Patent Classification (IPC)	02 March 2000 (02.03.2000) or national classification and IPC		UZ MIAFCH 1999 (UZ.U3.1999)		
IPC(7): C23C 22/07 and US Cl.: 148/26	:a				
Applicant 146/20					
HENKEL CORPORATION					
This international preliming	nary examination report has bee				
2. This REPORT consists of	a total of 5 sheets, including	this cover sheet			
which have been ame	ended and are the basis for this	report and/or sh	escription, claims and/or drawings eets containing rectifications made istrative Instructions under the PCT).		
These annexes consist of a	a total of sheets.				
3. This report contains indica	ations relating to the following	tems:			
I Basis of the rep	I Basis of the report				
II Priority					
III Non-establishm	ent of report with regard to nov	elty, inventive s	step and industrial applicability		
IV Lack of unity o	f invention				
V Reasoned stater	nent under Article 35(2) with re	egard to novelty	, inventive step or industrial		
ا لاستا	tations and explanations suppor				
VI Certain docume	ents cited				
VII Certain defects in the international application					
VIII Certain observa	tions on the international applic	cation			
Date of submission of the demand	Date	of completion o	of this report		
23 June 2000 (23.06.2000)	08 М	arch 2002 (08.03.	2002)		
Name and mailing address of the IPEA/		orized officer	. 90		
Commissioner of Patents and Trademar Box PCT	Andr	ew L Oltmans	Jean Proctor Paralegal S <sub>e</sub> ctualist		
Washington, D.C. 20231 Facsimile No. (703)305-3230		hone No. 703-30	8-0661		

Form PCT/IPEA/409 (cover sheet)(July 1998)



International application No.
PCT/US00/05458

I.	Basis	s of the report
1.	With	regard to the elements of the international application:*
	$\boxtimes$	the international application as originally filed.
	$\overline{\boxtimes}$	the description:
		pages 1-11 as originally filed
		pages NONE , filed with the demand
		pages NONE , filed with the letter of
	$\boxtimes$	the claims:
		pages 11 and 12 , as originally filed
		pages NONE, as amended (together with any statement) under Article 19 pages NONE, filed with the demand
		pages NONE , filed with the letter of
	Ш	the drawings: pages NONE , as originally filed
		pages NONE, filed with the demand
		pages NONE , filed with the letter of
		the sequence listing part of the description:
		pages NONE , as originally filed
		pages NONE , filed with the demand
		pages NONE, filed with the letter of
2.	With	regard to the language, all the elements marked above were available or furnished to this Authority in the
	langu	age in which the international application was filed, unless otherwise indicated under this item.  e elements were available or furnished to this Authority in the following language which is:
	THES	
	$\vdash$	the language of a translation furnished for the purposes of international search (under Rule23.1(b)).
	$\square$	the language of publication of the international application (under Rule 48.3(b)).
		the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).
3.	With	regard to any nucleotide and/or amino acid sequence disclosed in the international application, the
	inter	national preliminary examination was carried out on the basis of the sequence listing:
		contained in the international application in printed form.
		filed together with the international application in computer readable form.
		furnished subsequently to this Authority in written form.
		furnished subsequently to this Authority in computer readable form.
		The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the
		international application as filed has been furnished.
		The statement that the information recorded in computer readable form is identical to the written sequence listing
		has been furnished.
4.		The amendments have resulted in the cancellation of
		the description, pages NONE
		the claims, Nos. NONE
		the drawings, sheets/fig NONE
5.		This report has been established as if (some of) the amendments had not been made, since they have been considered to go
		beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).**
th	is repo	cement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in ort as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17). replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.
·	Any I	comment once containing once untertained once of experience to the containing once of



International application No. PCT/US00/05458

V. Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement				
1. STATEMENT				
Novelty (N)	Claims	1-11		YES
, , , , , , , , , , , , , , , , , , ,				NO
Inventive Step (IS)		NONE		YES
	Claims	1-11	·	NO
Industrial Applicability (IA)	Claims	1-11		YES
madstrial repplications (111)		NONE		NO
2. CITATIONS AND EXPLANATIONS Please See Continuation Sheet				
	•			
				•
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	,			

Form PCT/IPEA/409 (Box V) (July 1998)





International application No. PCT/US00/05458

Supplemental Box (To be used when the	space in any of the preceding boxes is not sufficient)
LACK OF INVEN	TIVE STEP al. U.S. Patent No. 5,703,733
5,703,733 (SPECK	-11 lack an inventive step under PCT Article 33(3) as being obvious over SPECKMANN et al. U.S. Patent No. (MANN).
SPECKM	MANN teaches a zinc phosphating solution having the following composition, which overlaps the composition, positional equations, recited in instant claims 1-6 (column 2):
including the comp	a) phosphating solutions containing the following com-
50	ponents are used:
	$Zn^{2+}$ cations in quantities of 0.1 to 5 g/l,
	PO <sub>4</sub> <sup>3—</sup> anions in quantities of 5 to 50 g/l,
	NO <sub>3</sub> anions in quantities of 0.1 to 50 g/l and
55	$Mn^{2+}$ cations in quantities of 0.1 to 5 g/l and
	Cu <sup>2+</sup> cations in quantities of 0.001 to 1 g/l,
SPECKMANN als	o teaches the addition of fluorine compounds, as recited in instant claim 7 (column 4, lines 47-49). SPECKMANN ocess of coating metal substrates wherein the conditions overlap those recited in instant claims 8-10 (column 2):
also teaches the pr	b) the following conditions are established: pH value of
	the phosphating solutions 1.5 to 4.5, temperature of the
	phosphating solutions 10° to 80° C., treatment time 1 to
60	300 seconds,

c) the workpieces are cathodically treated during phosphating with a direct current having a density of 0.01 to

 $100 \text{ mA/cm}^2$ .

[emphasis added by examiner]

SPECKMANN teaches that the process includes contacting the liquid composition with a counter electrode and causing an electric current to flow through the metal substrate and into the volume of the liquid composition, as recited in instant claim 8 (column 3, lines 45-50). SPECKMANN further teaches a pretreatment with the composition recited in instant claim 11 (column 5, lines 34-41 and



International application No.

55

PCT/US00/05458

Supplemental Box

(To be used when the space in any of the preceding boxes is not sufficient)

column 7, lines 26-30).

SPECKMANN fails to meet all of the limitations of the instant claims in that SPECKMANN fails to explicitly teach the exact compositional and process condition ranges instantly claimed.

However, one of ordinary skill in the art would find the instant invention obvious because the compositional ranges and the process conditions taught in SPECKMANN overlap the compositional ranges recited in the instant claims.

KANAMARU et al. U.S. Patent No. 5.525,431

Claims 1-10 lack an inventive step under PCT Article 33(3) as being obvious over KANAMARU et al. U.S. Patent No. 5.525.431 (KANAMARU).

KANAMARU teaches an electrolyte composition for electrolytically coating metal substrates wherein the electrolyte contains phosphoric acid, nitric acid and dissolved zinc cations in a concentration that overlaps the concentration, including the compositional equations, recited in instant claims 1-7 (column 15):

Such oxide film can be prepared, for example, by dipping zinc-base galvanized sheet steel in an aqueous solution containing 1-70 g/l of potassium permanganate, 5-60 g/l of phosphoric acid or boric acid (when the two acids are used together, respectively 5-60 g/l) and 100-800 g/l of zinc nitrate, by subjecting the galvanized sheet steel to a cathode electrolytic treatment in said aqueous solution, or by spraying the aqueous solution onto the galvanized sheet steel, whereby Mn oxide, phosphoric acid and Zn oxide are formed simultaneously.

[emphasis added by examiner] and

An etching agent, for example, at least one of sulfuric acid, nitric acid, perchloric acid, etc. is preferably added to the above-mentioned aqueous solution in an amount of 1–10 g/l to improve the adhesive property, etc. of the film.

[emphasis added by examiner] (see also column 18, lines 19-51)

KANAMARU also teaches a process of coating metal substrates wherein the process conditions overlap those recited in the instant claims (column 18, line 25 and 50). KANAMARU teaches that the process includes contacting the liquid composition with a counter electrode and causing an electric current to flow through the metal substrate and into the volume of the liquid composition, as recited in instant claim 8 (column 20, line 63 to column 23, line 3 and column 22, lines 28-35).

KANAMARU fails to meet all of the limitations of the instant claims in that KANAMARU fails to explicitly teach the exact compositional and process condition ranges instantly claimed.

However, one of ordinary skill in the art would find the instant invention obvious because the compositional ranges and the process conditions taught in KANAMARU overlap the compositional ranges recited in the instant claims.

#### INDUSTRIAL APPICABILITY

Claims 1-11 meet the requirement as defined by PCT Article 33(2) and 33(4) because the liquid composition and the process for forming a zinc phosphate conversion coating find use in the metal finishing industry.
NEW CITATIONS

Ner Dorch Ret PCT - 6/16/00
PCT PATENT COOPERATION TREATY From the INTERNATIONAL SEARCHING AUTHORITY To: HARPER, STEPHEN D. HENKEL CORPORATION 2500 RENAISSANCE BOULEVARD NOTIFICATION OF TRANSMITTAL OF SUITE 200 **GULPH MILLS, PA 19406** THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION Rev 1 DS/ISR - 6/16/00 (PCT Rule 44.1) Rude 120/15R-7/16/00 Jule 105/15R-8/16/00 Date of Mailing (day/month/year) **16** MAY 2000 Applicant's or agent's file reference See paragraphs 1 and 4 below FOR FURTHER ACTION M 6712 HST/NI International filing date International application No. (day/month/year) 02 MARCH 2000 PCT/US00/05458 Applicant HENKEL CORPORATION The applicant is hereby notified that the international search report has been established and is transmitted herewith. 1. X Filing of amendments and statement under Article 19: The applicant is entitled, if he so wishes, to amend the claims of the international application (see Rule 46): The time limit for filing such amendments is normally 2 months from the date of transmittal of the international search report; however, for more details, see the notes on the accompanying sheet international search report; however, for more details, see the notes on the accompanying sheet. Where? Directly to the International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35 REC'D MAY 1 7 2008 For more detailed instructions, see the notes on the accompanying sheet UE FILE M67/ The applicant is hereby notified that no international search report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith. Article 17(2)(a) to that effect is transmitted herewith. With regard to the protest against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that: the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices. no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made. The applicant is reminded of the following: 4. Further action(s): Shortly after 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in rules 90 bis 1 and 90 bis 3, respectively, before the completion of the technical preparations for international publication. Within 19 months from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later). Within 20 months from the priority date, the applicant must perform the prescribed acts for entry into the national phase

Name and mailing address of the ISA/US	Authorized officer Jam Back La
Commissioner of Patents and Trademarks Box PCT	ANDREW L OLTMANS
Washington, D.C. 20231	m 1 1 N (700) 000 0001
Faccimile No. (703) 305-3230	Telephone No. (703) 308-0661

priority date or could not be elected because they are not bound by Chapter II.

before all designated Offices which have not been elected in the demand or in a later election within 19 months from the

Form PCT/ISA/220 (July 1998) \*

(See notes on accompanying sheet)

## PATENT COOPERATION TREATY

## **PCT**

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

				I Banarah Banar
Applicant's or agent's file reference M 6712 HST/NI	FOR FURTHER ACTION	(Form PCT/ISA/220	)) as well as, wher	aternational Search Report e applicable, item 5 below.
International application No.	International filing date	e (day/month/year)	(Earliest) Priority	y Date (day/month/year)
PCT/US00/05458	02 MARCH 2000		02 MARCH	1999
Applicant HENKEL CORPORATION				
This international search report has been according to Article 18. A copy is being	ng transmitted to the Inter	national Bureau.	uthority and is tra	ansmitted to the applicant
This international search report consist	s of a total of 🖊 sheet	s.		
X It is also accompanied by a	copy of each prior art do	cument cited in this	report.	
Authority (Rule 23.1(b)).  b. With regard to any nucleotide was carried out on the basis of contained in the internation filed together with the internation furnished subsequently to the statement that the subsequently the statement the statement that the subsequently the statement that the subsequently the statement that the subsequently the statem	and/or amino acid sequence is carried out on the basis and/or amino acid sequence fithe sequence listing:  all application in written is contained application in contained and application in computer in this Authority in written in this Authority in computer in the computer is filed has been furnished written action recorded in computer in the contained in computer in the contained in the conta	of a translation of the ince disclosed in the ince disclosed in the inform.  In readable form.  In readable form.  In readable form.  In readable form is ideal.  In readable form is ideal.	he international applicanternational applicant	application furnished to this cation, the international search
5. With regard to the abstract,				
X the text is approved as su			••	•
the text has been establish Box III. The applicant ma search report, submit con	y, within one month from nments to this Authority.	the date of mailing	of this internation	in nal
6. The figure of the drawings to be	published with the abstr	act is Figure No		
as suggested by the applic				None of the figures.
because the applicant faile	ed to suggest a figure.			
because this figure better	characterizes the inventi	on.		

#### NOTES TO FORM PCT/ISA/220 (c ntinued)

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

- [Where originally there were 48 claims and after amendment of some claims there are 51]:
  "Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers;
  claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
- [Where originally there were 15 claims and after amendment of all claims there are 11]:
   "Claims 1 to 15 replaced by amended claims 1 to 11."
- [Where originally there were 14 claims and the \*mendments consist in cancelling some claims and an adding new claims]:
   "Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or "Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
- 4. [Where various kinds of amendments are made]:
  "Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into arcended claims 15, 16 and 17; new claims 20 and 21 addeed."

#### "Statement under Article 19(1)" (Rule 46,4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

The statement should be brief, it should not exceed 500 words if in English or if translated into English.

It should not be confounded with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It should not contain any disparaging comments on the international search report or the relevance of critations contained in that report. Reference to critations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

#### In what language?

The amendments must be made in the language in which the international application is published. The letter and any statement accompanying the amendments must be in the same language as the international application if that language is English or French; otherwise, it must be in English or French, at the choice of the applicant.

#### Consequence if a demand for international preliminary examination has already been filed?

If, at the time of filing any amendments under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the same time of filing the amendments with the International Bureau, also file a copy of such amendments with the International Preliminary Examining Authority (see Rule 62.2(a), first sentence).

#### Consequence with regard to translation of the international application for entry into the national planse?

The applicant's attention is drawn to the fact that, where upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, r in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see Volume II of the PCT Applicant's Guide.

63.02 g/mol × 0.2 mol/L	0.144 mol/L	Zn 65.38 g/mol	H3PO4 98 g/mol × 0.2 mol/L
12.604 g/L	9.41472 g/L		19.6 g/L
upper limit lower limit	Zn 65.38 g/mol	upper limit lower limit	Total Equation Inpu H3F
0.16 mol/L 0.144 mol/L	mol	H3PO4 nitric acid 0.06 0. 0.054 0.0	ut 904 nitrio
10.4608 g/L 9.41472 g/L		acid 0.1 0.16 0.09 0.144	e acid 0.2

0.6 mol/L	63.02 g/mol x	nitric acid	0.144 mol/L	×	65.38 g/mol	Zn			0.6 mol/L	×	98 g/mol	H3PO4
37.812 g/L			9.41472 g/L						58.8 g/L			
	upper limit	65.38 g/mol ×	Zn		lower limit	upper limit	_			_	_	Total Equation
C. TOP	0.48 mol/L	g/mol ×				0.18 0.3	H3PO4 nitric acid	Calculated Zn conc	0.6 0.6	H3PO4 nitric acid	Input	on
70.644	31.3824 g/L				_	3 0.48			0	_		

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cess of this invention enables zinc phosphate treatment to be run very rapidly through the use of electrolysis. This feature, in combination with the fact that this process can be used to execute zinc phosphate treatment on essentially any material that is electrically conductive, makes the instant process highly advantageous on an industrial or commercial basis.

(

#### **CLAIMS**

1. A liquid composition of matter that is suitable as electrolyte for a nonsludging electrolytic zinc phosphate treatment process, said liquid composition comprising water, dissolved phosphoric acid, dissolved nitric acid, dissolved zinc cations, m chemically distinct species of cations other than zinc, and n chemically distinct species of anions other than anions derivable by ionization of phosphoric and nitric acids, each of m and n independently being zero or a positive integer, the concentration of zinc in moles per liter in said liquid composition satisfying the following mathematical condition:

$${\rm Zn} \le 0.3 {\rm H_3PO_4} + 0.5 {\rm HNO_3} - 0.5 \sum_{i=0}^{m} p_i C_i + 0.5 \sum_{i=0}^{n} q_i A_i$$

in which: " $\{Zn\}$ ", " $\{H_3PO_4\}$ ", and " $\{HNO_3\}$ " respectively represent the zinc, phosphoric acid, and nitric acid concentrations in mol/L; each of  $C_0$  and  $A_0$  is zero; each of  $p_0$  and  $q_0$  is 1; if m is not zero, for each positive integer i from 1 to m,  $C_i$  represents the concentration in mol/L of the ith distinct cation species other than zinc present in the bath and  $p_i$  represents the cationic valence of said ith distinct cation species; and if n is not zero, for each positive integer j from 1 to n,  $A_j$  represents the concentration in mol/L of the jth distinct anion species other than anions derivable by ionization of phosphoric or nitric acids present in the bath and  $q_j$  represents the anionic valence of said jth distinct anion species.

- 2. A liquid composition according to claim 1, wherein:
- the phosphoric acid concentration is from 0.10 to 0.60 mol/L;
- the nitric acid concentration is from 0.20 to 1.0 mol/L; and

<sup>30</sup> - 
$$\{Zn\} \ge 0.15 \{H_3PO_4\} + 0.25 \{HNO_3\} - 0.25 \sum_{i=0}^{m} p_i C_i + 0.25 \sum_{i=0}^{n} q_i A_i$$

- 3. A liquid composition according to claim 2, wherein:
- the phosphoric acid concentration is from 0.25 to 0.50 mol/L;
- the nitric acid concentration is from 0.65 to 0.90 mol/L; and

$${\rm Zn} \ge 0.27 {\rm H}_3{\rm PO}_4 + 0.45 {\rm HNO}_3 - 0.45 \sum_{i=0}^m p_i C_i + 0.45 \sum_{j=0}^n q_j A_j$$

4. A liquid composition according to claim 3, wherein  $\{Zn\}/\{H_3PO_4\} < 0.91$ .

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5. A liquid composition according to claim 2, wherein  $\{Zn\}/\{H_3PO_4\} < 0.91$ .

- 6. A liquid composition according to claim 1, wherein  $\{Zn\}/\{H_3PO_4\} < 0.91$ .
- 7. A liquid composition according to any one of claims 1 through 6, additionally comprising at least one additive selected from the group consisting of nitrous acid, permanganic acid, peroxysulfuric acid, hydrogen peroxide, chloric acid, perchloric acid, nitrobenzene sulfonic acid, hydroxylamine, starch/phosphoric acid esters, fluorine compounds, and salts of all of the other materials previously recited in this group for which salts are known.
- 8. A process for forming a zinc phosphate conversion coating on a metal substrate without generating any sludge thereby, said process comprising operations of:
- bringing said metal substrate into contact with a volume of a liquid composition according to any one of claims 1 through 7, said volume of liquid composition also being in contact with a counter electrode that is distinct from said metal substrate; and
- causing electric current to flow in a cathodizing direction through said metal substrate into said volume of liquid composition and through said counter electrode.
  - 9. A process according to claim 8, wherein:
  - said volume of liquid composition is maintained during operation (II) at a temperature that is between 50 and 85 °C; and
- in operation (II) there is a current density through said metal substrate that is between 0.5 and 50 A/dm².
  - 10. A process according to claim 9, wherein:
  - said volume of liquid composition is maintained during operation (II) at a temperature that is between 75 and 85 °C; and
- in operation (II) there is a current density through said metal substrate that is between 7.0 and 15 A/dm².
  - 11. A process according to any one of claims 8 through 10, wherein prior to operation (I), said metal substrate is brought into contact with a weakly basic aqueous colloidal solution that contains titanium oxide, titanium hydroxide, and zinc phosphate.

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add A57

#### IN THE CLAIMS:

#### Amend claims 7, 8 and 11 to read as follows:

- A3
- 7. (Amended) A liquid composition according to claim 1, additionally comprising at least one additive selected from the group consisting of nitrous acid, permanganic acid, peroxysulfuric acid, hydrogen peroxide, chloric acid, perchloric acid, nitrobenzene sulfonic acid, hydroxylamine, starch/phosphoric acid esters, fluorine compounds, and salts of all of the other materials previously recited in this group for which salts are known.
- 8. (Amended) A process for forming a zinc phosphate conversion coating on a metal substrate without generating any sludge thereby, said process comprising operations of:
  - (I) bringing said metal substrate into contact with a volume of a liquid composition according to claim 1, said volume of liquid composition also being in contact with a counter electrode that is distinct from said metal substrate; and
  - (II) causing electric current in flow in a cathodizing direction through said metal substrate into said volume of liquid composition and through said counter electrode.
- A4

11. (Amended) A process according to claim 8, wherein prior to operation (I), said metal substrate is brought into contact with a weakly basic aqueous collodial solution that contains titanium oxide, titanium hydroxide, and zinc phosphate.

#### Enter new claims 12-21 as follows:



--12. (New) A liquid composition that is suitable as electrolyte for a nonsludging electrolytic zinc phosphate treatment process, said liquid composition comprising water, at least 0.10 mol/L dissolved phosphoric acid, at least 0.10 mol/L dissolved nitric acid, dissolved zinc cations, m chemically distinct species of cations other than zinc, and n chemically distinct species of anions other than anions derivable

by ionization of phosphoric and nitric acids, each of m and n independently being zero or a positive integer, the concentration of zinc in moles per liter in said liquid composition satisfying both of the following mathematical conditions:

$$\{Zn\} \le 0.3 \ \{H_3PO_4\} + 0.5 \ \{HNO_3\} - 0.5 \sum_{i=0}^{m} p_i C_i + 0.5 \sum_{j=0}^{n} q_j A_j$$
, and

$$\{Z_n\} \ge 0.15 \{H_3PO_4\} + 0.25 \{HNO_3\} - 0.25 \sum_{i=0}^{m} p_i C_i + 0.25 \sum_{j=0}^{n} A_j$$

in which: " $\{Zn\}$ ", " $\{H_3PO_4\}$ ", and " $\{HNO_3\}$ " respectively represent the zinc, phosphoric acid, and nitric acid concentrations in mol/L; each of  $C_0$  and  $A_0$  is zero; each  $p_0$  and  $q_0$  is 1; if m is not zero for each positive integer j from 1 to m,  $C_j$  represents the concentration in mol/L of the jth distinct cation species other than zinc present in the bath and  $p_j$  represents the cationic valence of said jth distinct cation species; and if n is not zero, for each positive integer j from 1 to n,  $A_j$  represents the concentration in mol/L of the jth distinct anion species other than anions derivable by ionization of phosphoric or nitric acids present in the bath and  $q_j$  represents the anionic valence of said jth distinct anion species, wherein  $\{Zn\}/\{H_3PO_4\}$  < 0.91.--

- --13. (New) A liquid composition according to claim 12, additionally comprising 0.0005 to 1.0 mol/L of at least one additive selected from the group consisting of nitrous acid, permanganic acid, peroxysulfuric acid, hydrogen peroxide, chloric acid, perchloric acid, nitrobenzene sulfonic acid, hydroxylamine, starch/phosphoric acid esters, fluorine compounds, and salts of all the other materials previously recited in this group for which salts are known.--
- --14. (New) A liquid composition of matter that is suitable as electrolyte for a nonsludging electrolytic zinc phosphate treatment process, said liquid composition comprising water, at least 0.20 mol/L dissolved

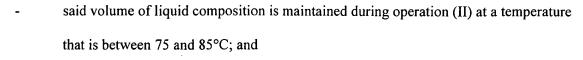
phosphoric acid, at least 0.20 mol/L dissolved nitric acid, dissolved zinc cations, m chemically distinct species of cations other than zinc, and n chemically distinct species of anions other than anions derivable by ionization of phosphoric and nitric acids, each of m and n independently being zero or a positive integer, the concentration of zinc in moles per liter in said liquid composition satisfying both of the following mathematical conditions:

$$\{Zn\} \leq 0.3 \ \{H_3PO_4\} \, + \, 0.5 \ \{HNO_3\} \, - \, 0.5 \, \sum_{i=0}^m \!\!\! p_i C_i \, + \, 0.5 \, \sum_{j=0}^n \!\!\! q_j A_j; \ \text{and}$$

in which: " $\{Zn\}$ ", " $\{H_3PO_4\}$ ", and " $\{HNO_3\}$ " respectively represent the zinc, phosphoric acid, and nitric acid concentrations in mol/L; each of  $C_0$  and  $A_0$  is zero; each  $p_0$  and  $q_0$  is 1; if m is not zero for each positive integer j from 1 to m,  $C_j$  represents the concentration in mol/L of the jth distinct cation species other than zinc present in the bath and  $p_j$  represents the cationic valence of said jth distinct cation species; and if n is not zero, for each positive integer j from 1 to n,  $A_j$  represents the concentration in mol/L of the jth distinct anion species other than anions derivable by ionization of phosphoric or nitric acids present in the bath and  $q_j$  represents the anionic valence of said jth distinct anion species, wherein  $\{Zn\}/\{H_3PO_4\}$  < 0.91.--

--15. (New) A liquid composition according to claim 14, additionally comprising 0.0005 to 1.0 mol/L of at least one additive selected from the group consisting of nitrous acid, permanganic acid, peroxysulfuric acid, hydrogen peroxide, chloric acid, perchloric acid, nitrobenzene sulfonic acid, hydroxylamine, starch/phosphoric acid esters, fluorine compounds, and salts of all the other materials previously recited in this group for which salts are known--

- --16. (New) A process for forming a zinc phosphate conversion coating on a metal substrate without generating any sludge thereby, said process comprising operations of:
  - (I) bringing said metal substrate into contact with a volume of a liquid composition according to claim 12, said volume of liquid composition also being in contact with a counter electrode that is distinct from said metal substrate; and
  - (II) causing electric current to flow in a cathodizing direction through said metal substrate into said volume of liquid composition and through said counter electrode--
- --17. (New) A process according to claim 16, wherein:
  - said volume of liquid composition is maintained during operation (II) at a temperature that is between 50 and 85°C; and
    - in operation (II) there is a current density through said metal substrate that is between 0.5 and 50 A/dm<sup>2</sup>.--
- --18. (New) A process according to claim 16, wherein prior to operation (I), said metal substrate is brought into contact with a weakly basic aqueous colloidal solution that contains titanium oxide, titanium hydroxide, and zinc phosphate--
- --19. (New) A process for forming a zinc phosphate conversion coating on a metal substrate without generating any sludge thereby, said process comprising operations of:
  - (I) bringing said metal substrate into contact with a volume of a liquid composition according to claim 14, said volume of liquid composition also being in contact with a counter electrode that is distinct from said metal substrate; and
  - (II) causing electric current to flow in a cathodizing direction through said metal substrate into said volume of liquid composition and through said counter electrode.--
- --20. (New) A process according to claim 19, wherein:



- in operation (II) there is a current density through said metal substrate that is between 7.0 and 15  $A/dm^2$ .--
- --21. (New) A process according to claim 19, wherein prior to operation (I), said metal substrate is brought into contact with a weakly basic aqueous colloidal solution that contains titanium oxide, titanium hydroxide, and zinc phosphate .--